Faster Tropical Upper Stratospheric Upwelling Drives Changes in Ozone Chemistry

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BOTTOM LINE: Upper Strat Composition Change →
O_3 Loss Changes → O_3 trends from 2005-2021

Strahan et al. (2022), Geophysical Research Letters

AMS, January 11, 2023

Quasi-Decadal (QD) N_2O Change over 2005-2021

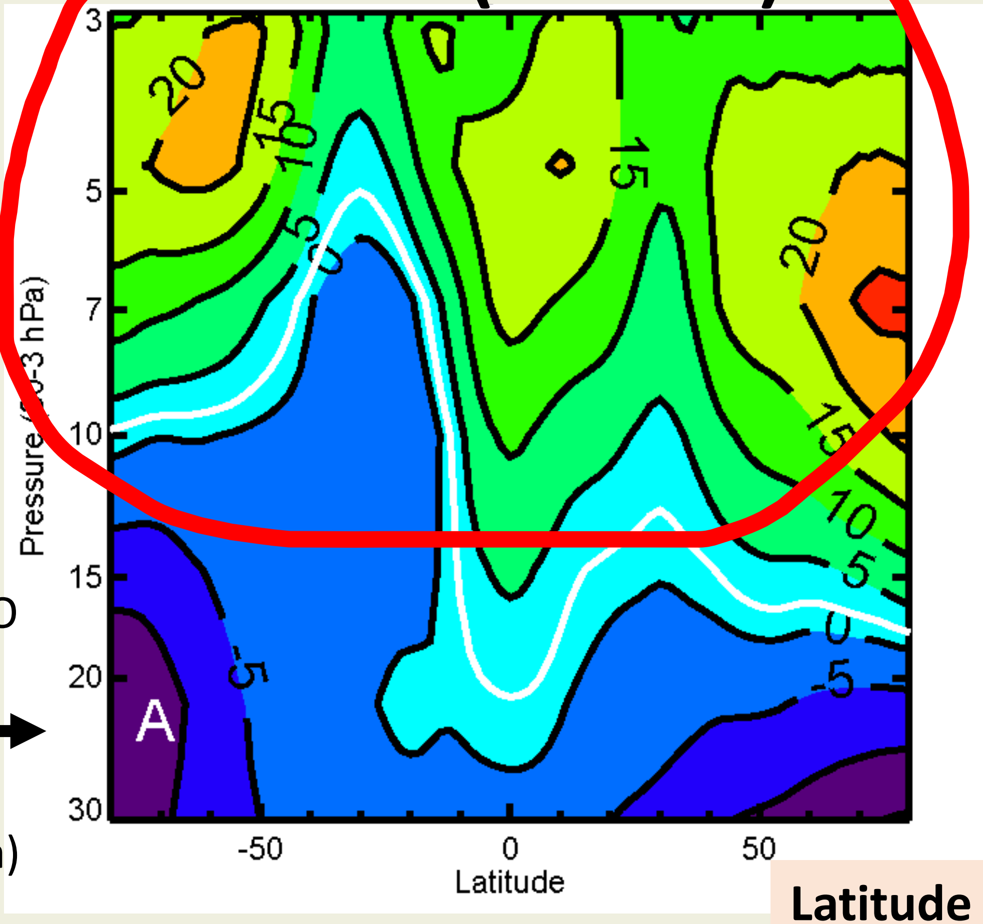
(*Percent change of 2013-2021 mean - 2005-2013 mean*)

N_2O surface growth over this period = 2%

No drift in MLS N_2O 22 hPa and above

Known negative drift below 22 hPa)

Obs (MLS V5)



Latitude

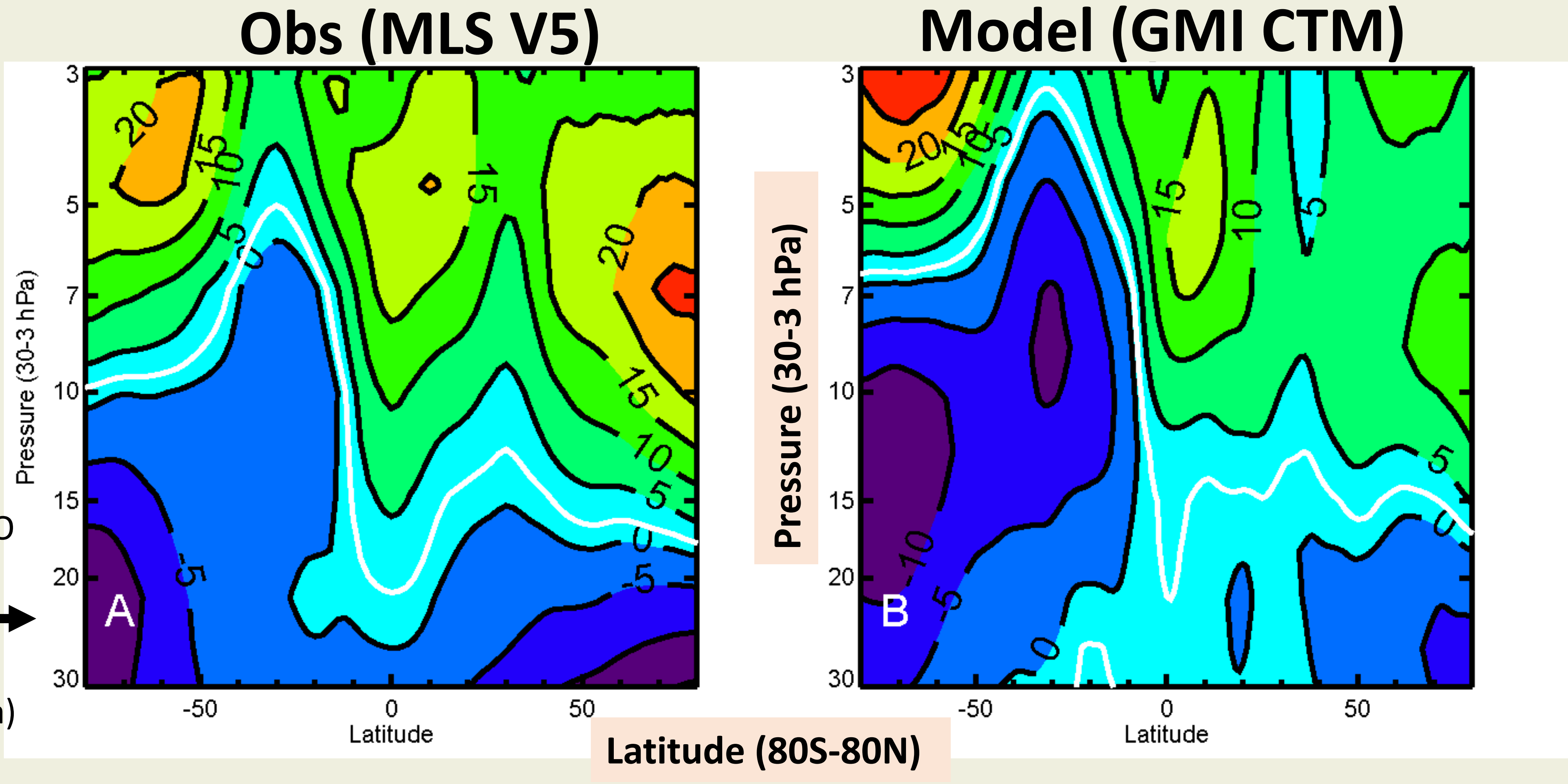
Quasi-Decadal (QD) N_2O Change over 2005-2021

(*Percent change of 2013-2021 mean - 2005-2013 mean*)

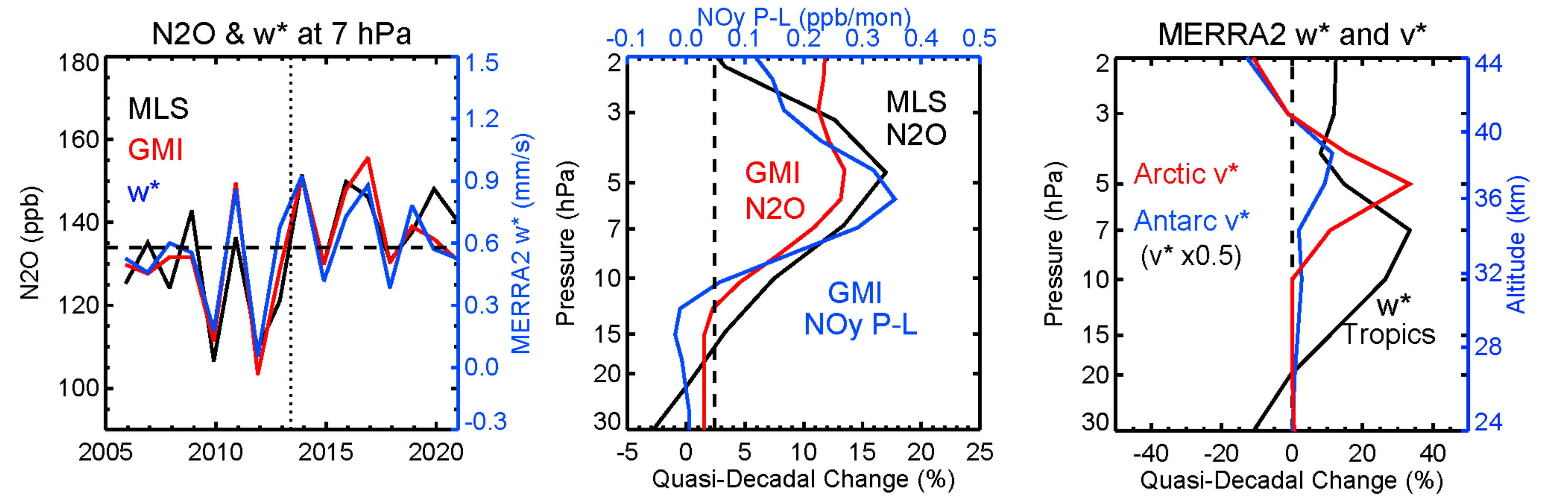
N_2O surface growth over this period = 2% (shown by white contour)

No drift in MLS N_2O 22 hPa and above

Known negative drift below 22 hPa)



Upper Stratospheric Circulation Change (w* and v*) Explains why is N_2O so high



Tropical N_2O grows above 10 hPa, leading to increased odd nitrogen production.

Upwelling (w*) increases above 20 hPa up to 30%

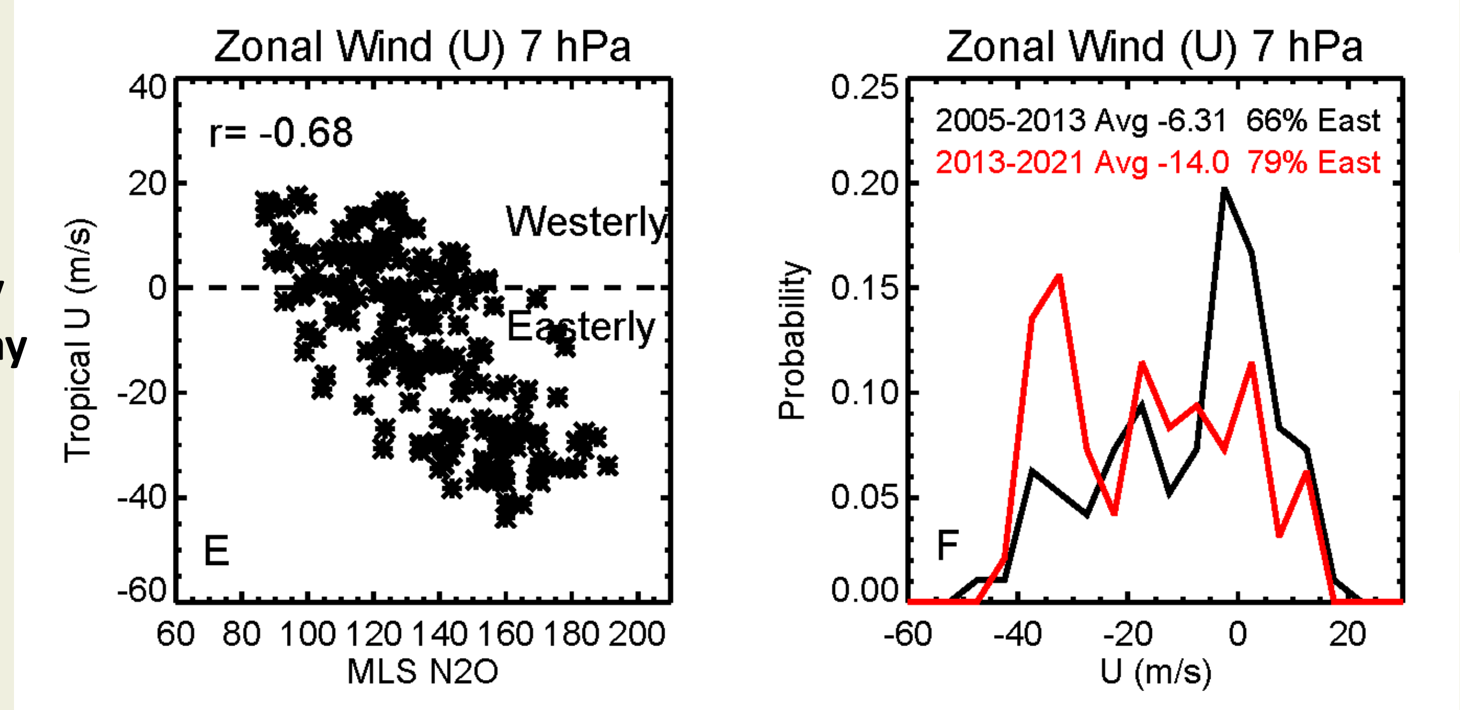
Arctic poleward transport (v* 50-70°N) increases by 60% at 5 hPa

The Quasi-Biennial Oscillation (QBO) modulates tropical upwelling: stronger during Easterlies, weaker in Westerlies

MLS tropical monthly mean N_2O is positively correlated with w* and anticorrelated with the zonal wind (i.e., QBO)

Stronger and more frequent QBO Easterlies 10-3 hPa increased mean tropical upwelling from 2013-2021

MLS and MERRA2 Monthly data from June 2005 to May 2021



2013-2021 was QBO-E 79% of the time

Two GMI Chemistry Transport Model (CTM) simulations with the same chemistry but different dynamics

GMI Chemistry Transport Model integrated with MERRA2:

- BASELINE: Time-varying MERRA2 Fields from June 2005-May 2021
- Fixed Dyn: June 2005-May 2007 Merra2 Fields (2 years) recycled until May 2021
- Both Simulations are forced with the same time-dependent source gases (N_2O, CFCs)

These start/end dates for the 2-year repeat chosen because of a 2-yr period QBO. This minimizes the transport adjustment when recycling.

O_3 loss differences between BASELINE and FIXED DYN show how *dynamical changes led to chemical changes* that affected O_3.

Compare QD Change in O_3 Loss from NO_x and ClO_x (ppb O_3/mo) in BASELINE and Fixed Dyn Simulations

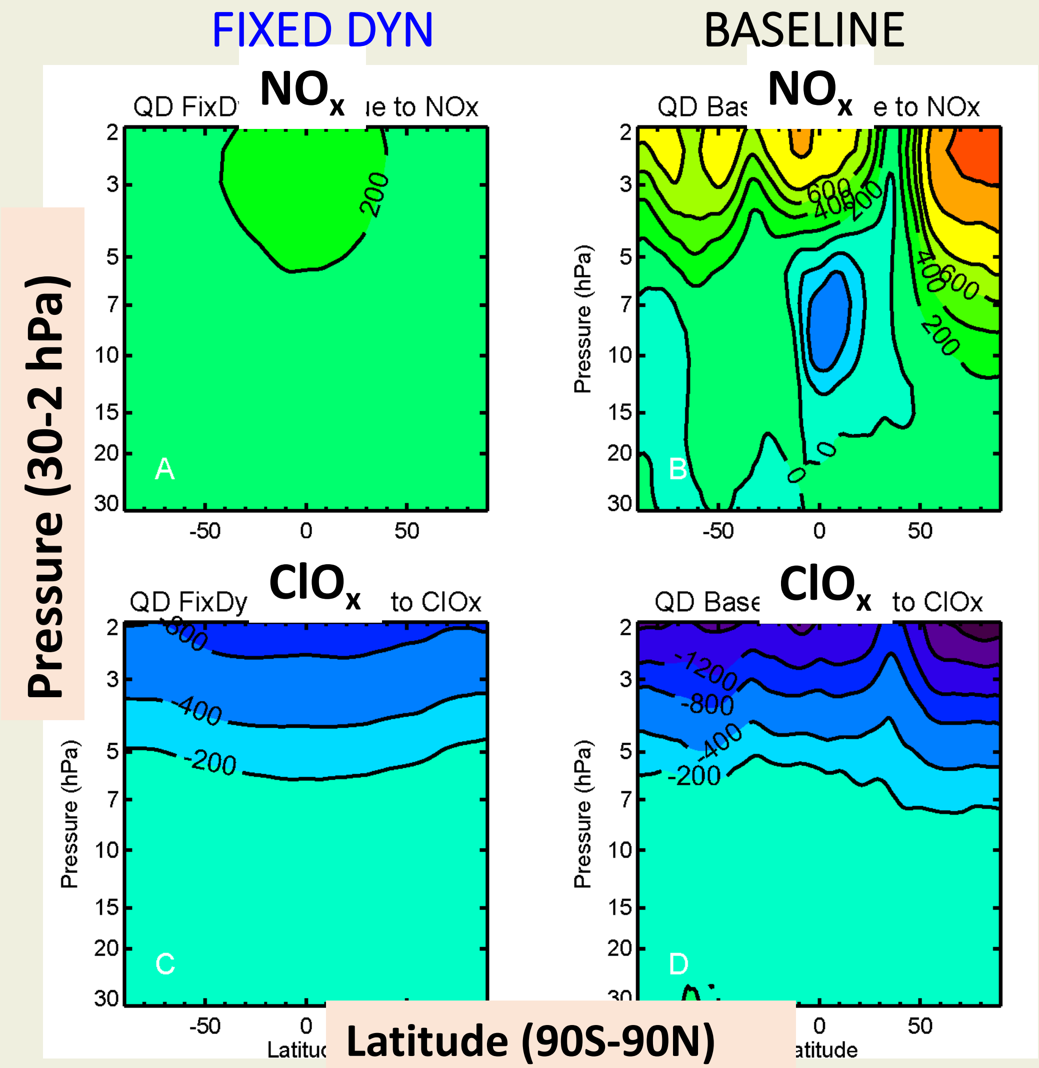
Fixed Dyn:

Hemispheric Symmetry

This is consistent with expectations from trends in tropospheric source gases (CFCs, N_2O)

Loss by NO_x increases above 5 hPa

Loss by ClO_x decreases with increasing altitude



BASELINE:

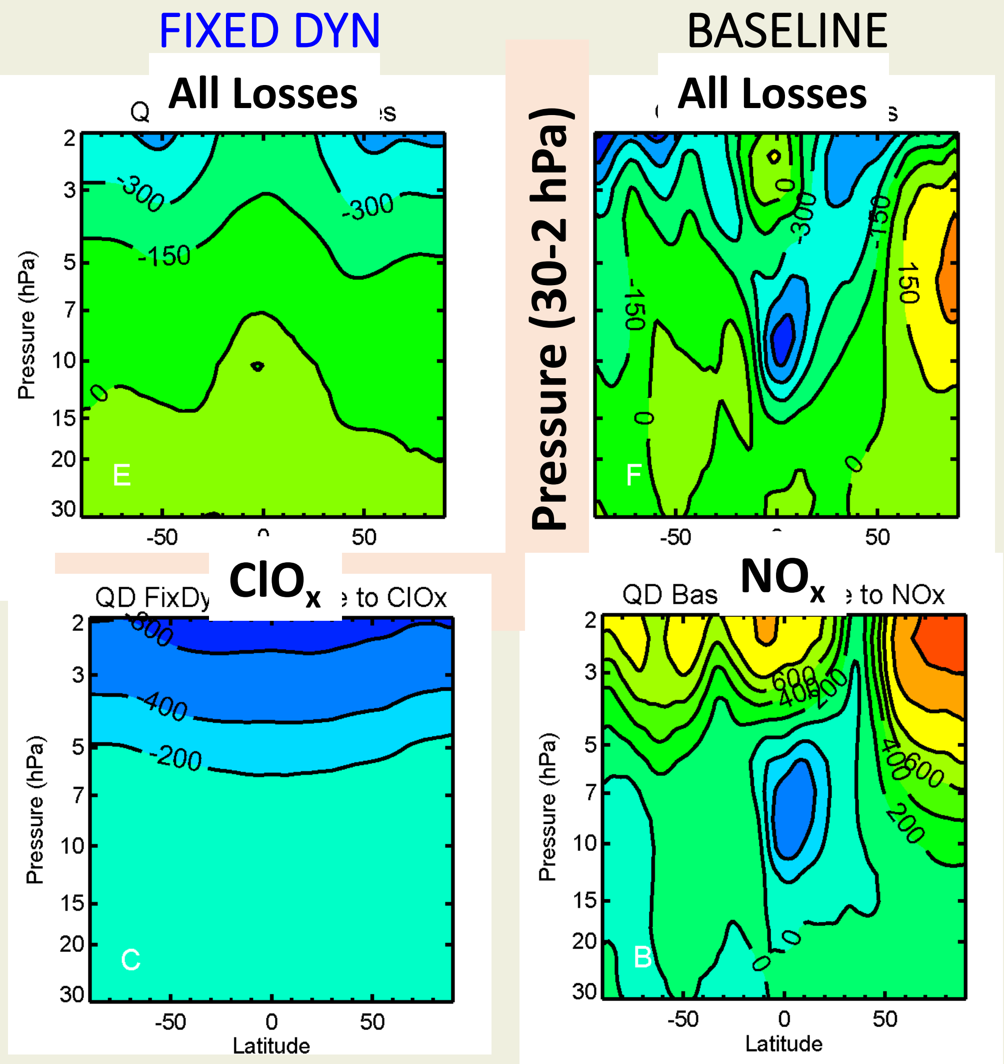
Hemispheric asymmetry

Greater loss by NO_x above 5 hPa, especially in the Arctic

Less loss by NO_x in the tropical middle strat

Loss by ClO_x decreases even more than FixedDyn in the Arctic Upper Strat

The QD Change in the Sum of all O_3 Losses (NOx, ClOx, HOx, etc.) is driven by the NO_x Loss Changes in Baseline



Fixed Dyn: QD O_3 loss changes are dominated by Cl changes

BASELINE:

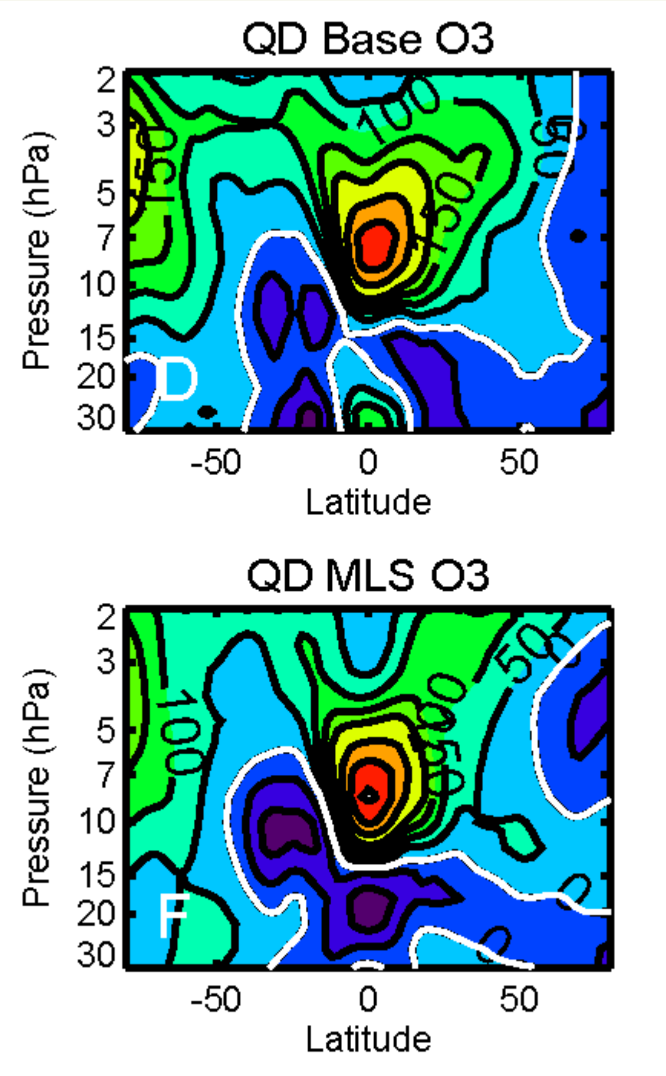
QD O_3 loss changes are dominated by NO_x changes. ClO_x changes contribute ~3 hPa and above.

And the denouement….BASELINE QD O_3 changes (ppb) look very much like observed O3 changes 2005-2021

<u>The Tropics</u>: middle strat has a big O_3 increase because increased upwelling reduced NO_x and the loss it causes

Arctic Upper Strat: O_3 decreases! It's not a lot (<200 ppb) but it's not increasing. Driven by NO_x

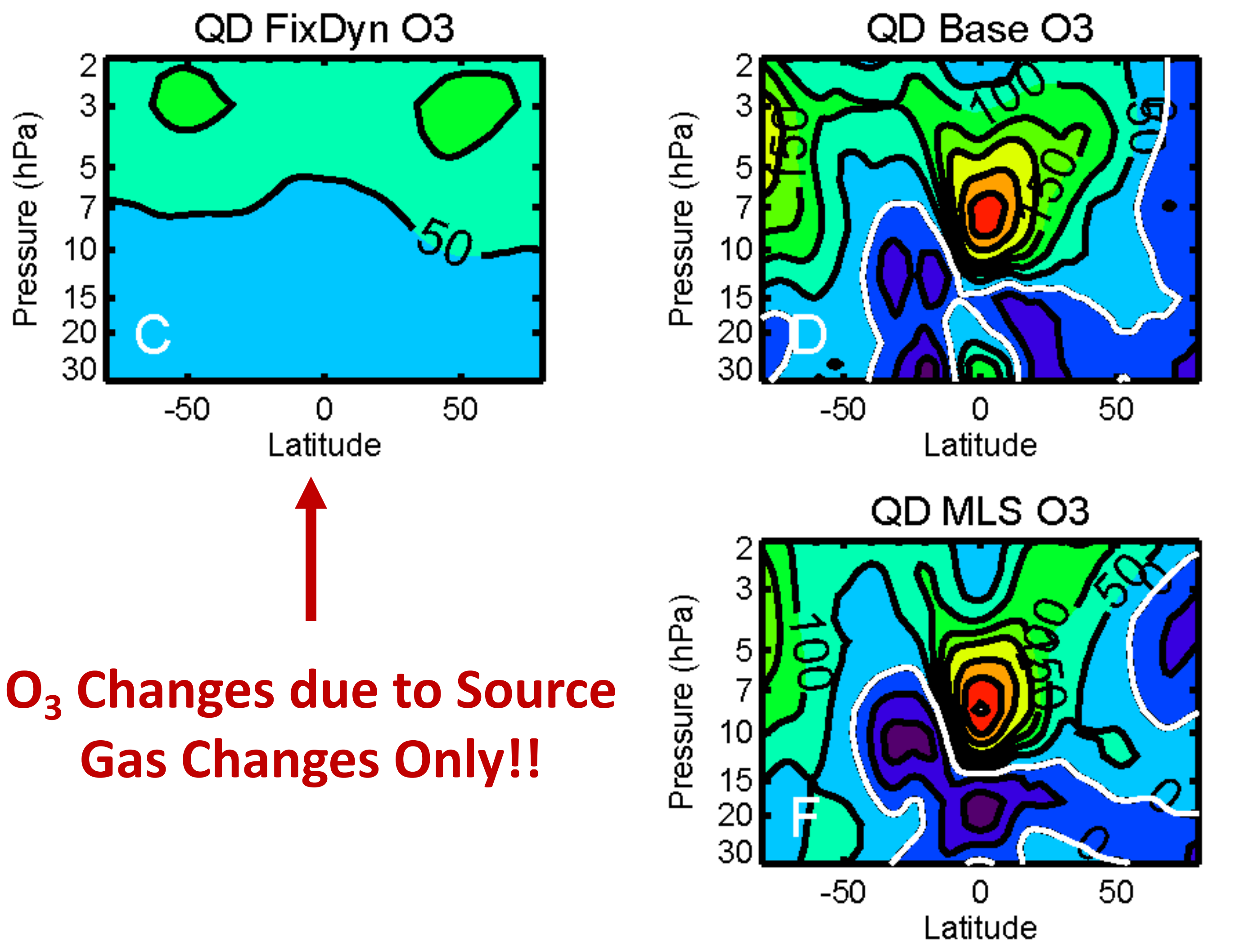
Antarctic Upper Strat: O_3 increases – reduced loss by ClO_x and less loss by NO_x (15-5 hPa)



And the denouement….Fixed Dyn QD O_3 changes look nothing like observed O3 changes 2005-2021

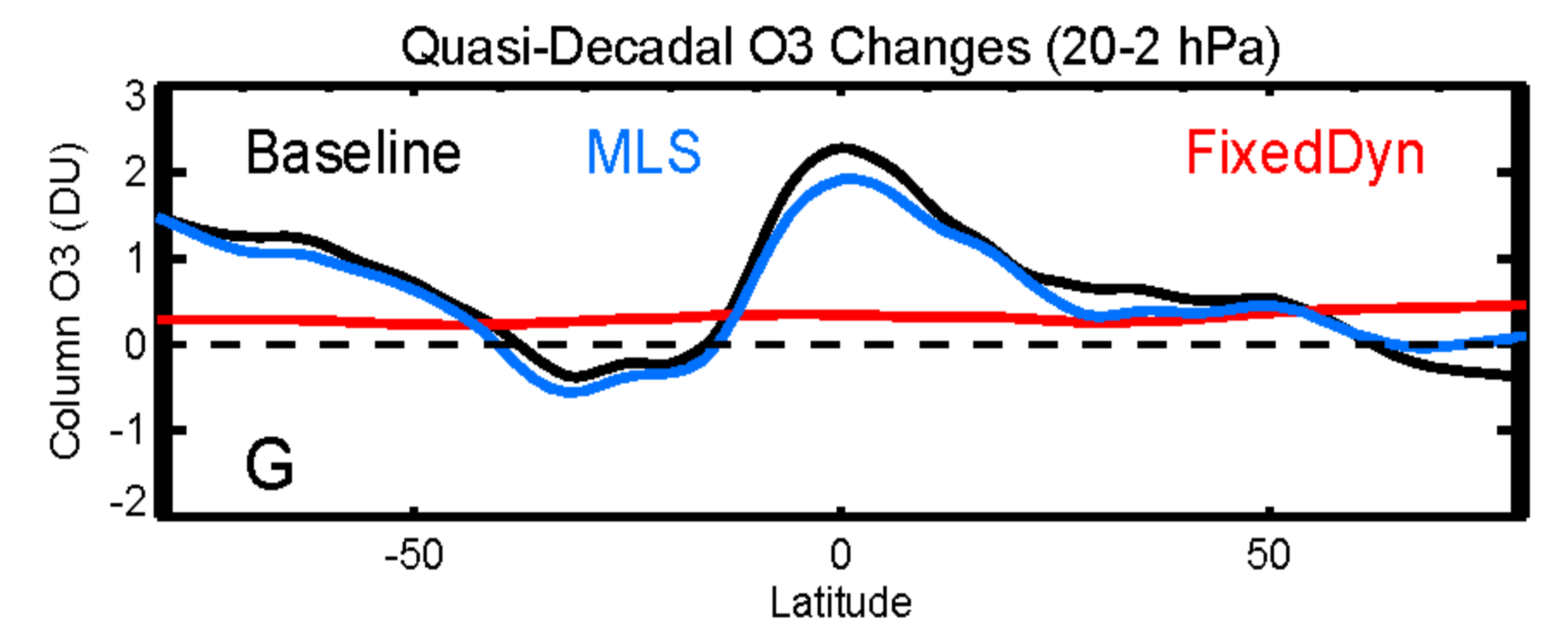
The Dynamically driven chemical changes are BASELINE – FixDyn O_3 (lower left panel).

Much of the observed O_3 changes 2005-2021 are caused by dynamically driven composition change!!



O_3 Changes due to Source Gas Changes Only!!

Dynamically driven chemical changes from 20-2 hPa column Affect Total Column O_3 Trends



Check out the excellent agreement between MLS (blue) and the GMI CTM (black)

The 20-2 hPa column O_3 Quasi-Decadal changes caused by dynamics:

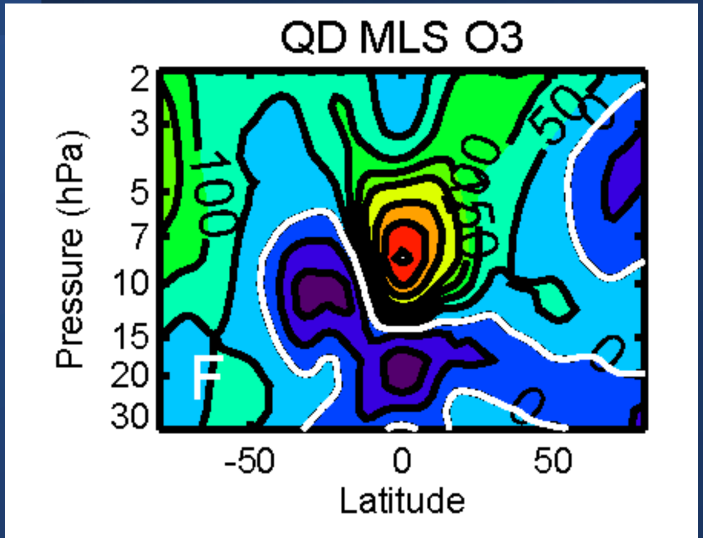
+2 DU in the tropics, -0.5 DU north of 60^{o}N, and +1 DU in the Antarctic.

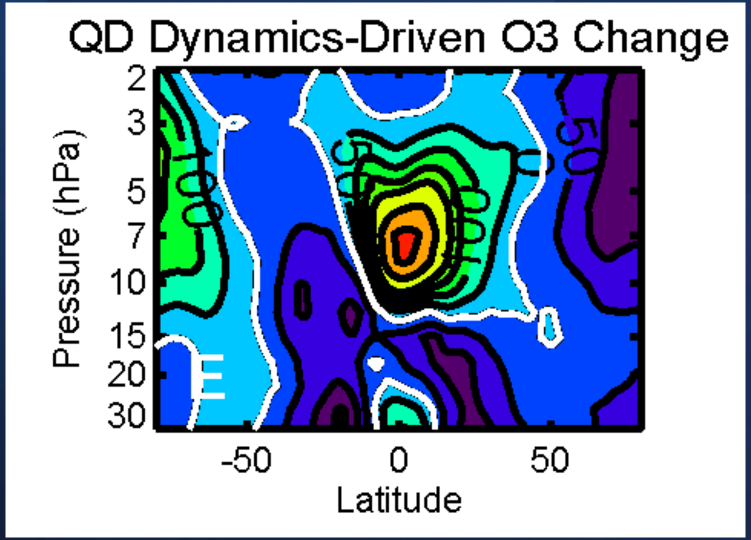
Questions/Thoughts…

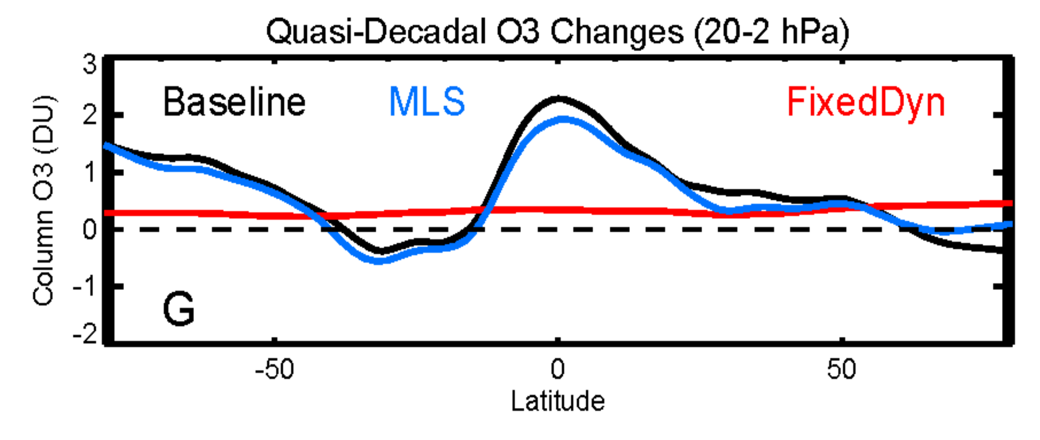
Will QBO changes persist? Is this a trend or just variability?

Chemistry Climate Models parameterize the QBO and can't respond physically to changes in forcing. They can't produce/predict this kind of effect on O_3.

O_3 trend regressions fit the QBO with terms for tshe QBO 30 and 50 hPa (transport!). This can't regress <u>changes in composition that affect O_3 chemistry</u>

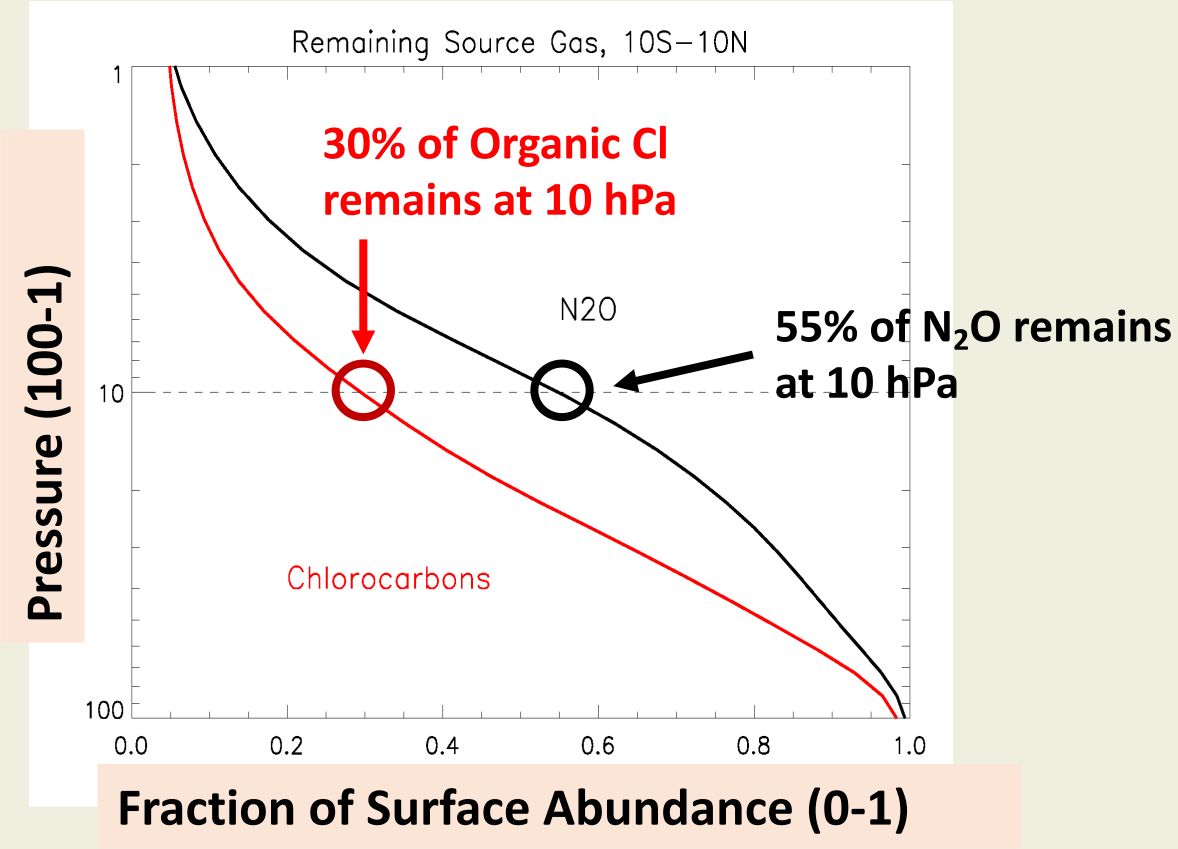






Please check out our 2022 GRL paper (Strahan et al.)

Why is there hemispheric asymmetry in Loss by NO_x but not the loss by ClO_x?

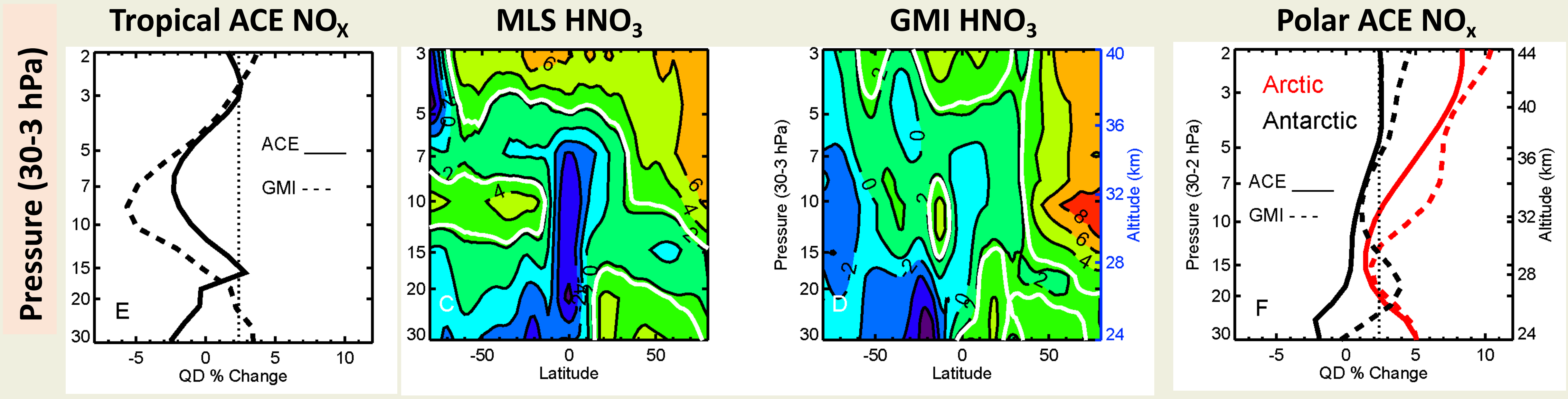


Upwelling changes affect product gases (e.g., radicals) by changing source gas distributions

The upwelling changes are 10 hPa and above

The CFCs are mostly photolyzed below 10 hPa but most N_2O is still unreacted.

Chemical impacts of Circulation Change: Increased Net Production of NO_y is transported to the Arctic

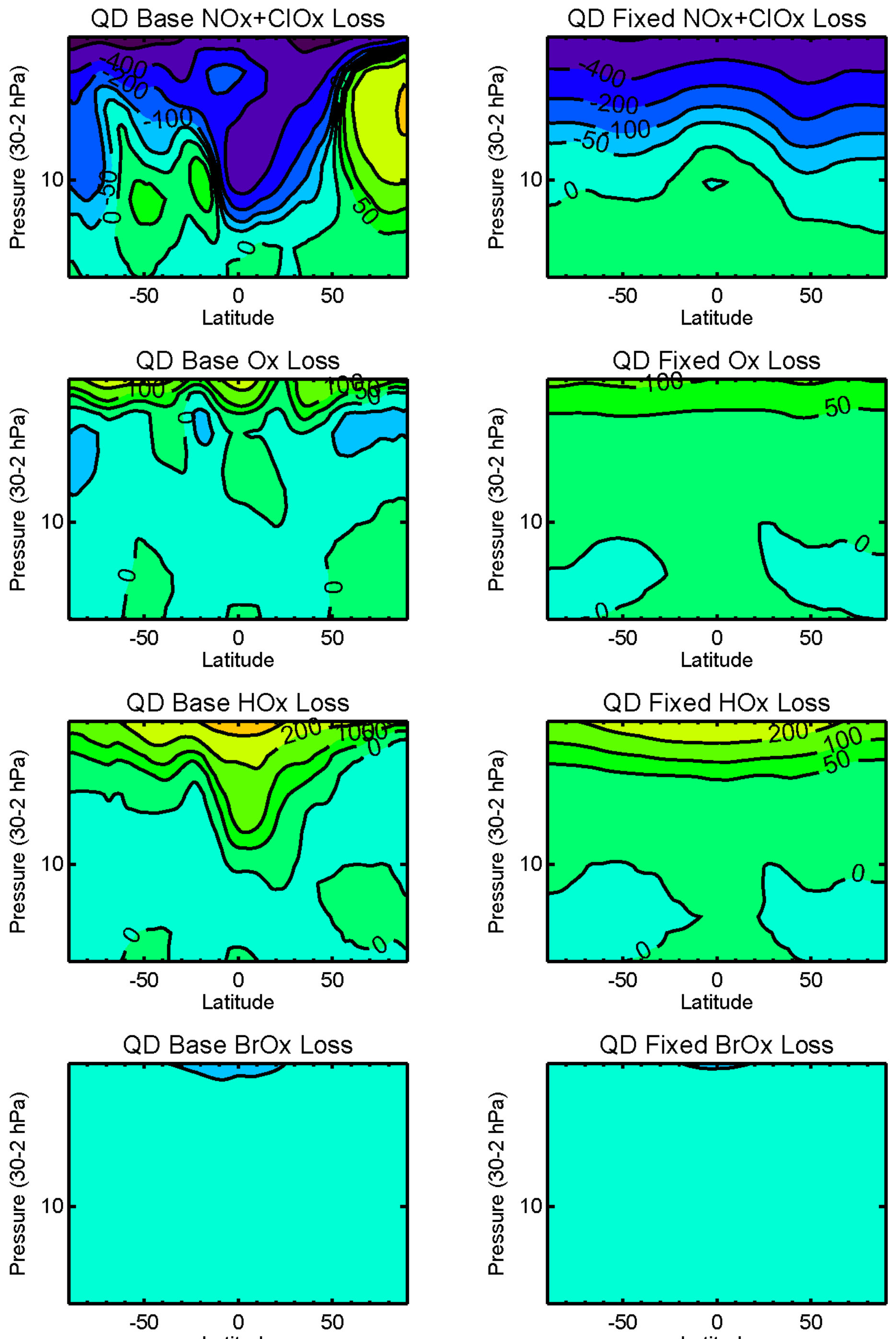


In the tropics, N_2O is relatively long-lived below ~7 hPa, thus N2O and NOy are largely transport controlled.

N_2O and NO_y are anti-correlated

All O3 Loss Cycles (QD Change), Fixed Dyn (left) and Baseline (right)

NOx + ClOx
Ox
HOx
BrOx



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